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Emission spectra of liquid-jet targets of hexane C_6H_{14}, dichloromethane CH_2Cl_2, methylene bromide CH_3Br in the range 4−20 nm under pulsed laser excitation

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The article discusses the results of studying the emission spectra of liquid hexane C_6H_{14}, dichloromethane CH_2Cl_2, methylene bromide CH_3Br upon excitation by pulsed laser radiation. We used a Nd:YAG,laser, $\lambda = 1064$ nm, $\tau = 5.2$ ns, $E_{\text{pulse}} = 0.8$ J. The spectral range of 4−20 nm was studied. We used injector with a hole 225 μm ?m in diameter, operating in a pulsed mode to form a liquid-droplet jet. The emission spectra of these liquid-jet targets were obtained. The obtained spectra were decoded, and the ions emitting in this spectral range were determined

Keywords: extreme ultraviolet radiation, emission spectra, laser spark, x-ray spectrometer-monochromator, liquid jets.

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Introduction

We have already examined the emission properties of gas-jet sources of extreme ultraviolet (EUV) and soft X-ray (SXR) radiation under pulsed laser excitation in [1–3]. While such sources are very convenient, they are characterized by low densities of matter in the laser spark generation region. Certain elements are also hard to introduce into the reaction volume of gas-jet laser-plasma radiation sources (LPSs), since gaseous compounds need to be used. An original system for introducing the studied substances into the reaction volume in the form of a pulsed liquid (droplet) jet was developed in order to solve this problem. This provided an opportunity to increase the density of matter in the laser spark generation region, expand the range of studied chemical elements, and reduce considerably the load on the evacuation system. In the present study, liquid jets of hexane C_6H_{14}, dichloromethane CH_2Cl_2, and methylene bromide CH_3Br were examined.

Experimental setup

The setup used in this research has been described in detail in [4]. It is operated as follows. The studied liquid is fed to a Bosch 0 280 158 017 injector installed within the evacuated volume. The nozzle has four apertures 225 μm in diameter on its end face. Three of them were sealed, and only the remaining one was used. This system is much easier to evacuate than a gas-jet radiation source; a cryopump cooled with liquid nitrogen is sufficient for this purpose. Laser radiation is directed to a short-focus lens, which focuses it to a plume of sprayed liquid. Laser-induced breakdown and plasma formation occur in this plume of liquid. Polychromatic SXR and EUV plasma radiation passes through an RSM-500 monochromator spectrometer and is detected by a pulsed detector.

An NL300 Series Nd:YAG laser, which was used to excite the gas jet, had the following parameters: a wavelength of 1064 nm, a laser pulse energy of 0.8 J, a pulse duration of 5.2 ns, and a pulse repetition rate of 10 Hz. A lens with a focal length of 45 mm is used to focus laser radiation to a liquid target. The calculated focal spot diameter is 66 μm. A freely suspended Mo/$ZrSi_2$ filter with layer thicknesses of 1.5/2.5 nm in a bilayer and 12 bilayers is installed at the RSM-500 input. This filter suppresses visible and VUV plasma radiation and stops particles of various nature forming in the course of operation of the SXR and EUV radiation source.

A spherical mirror ($R = 4$ m) and a grating ($R = 3$ m, 600 lines/mm) fabricated from glass K8 with gold coating were used in measurements with RSM-500. The studied wavelength range was 4−20 nm. The spectral resolution of the instrument measured at the L absorption edges of Si and Al and the K absorption edge of Be of freely suspended filters and determined based on the FWHM of the zeroth order was 0.04 nm.

Liquid-droplet jets forming in the process of outflow from the injector into vacuum normally have a complex spatial structure that is shaped by the thermodynamic properties and parameters of liquid fed through the injector. Several

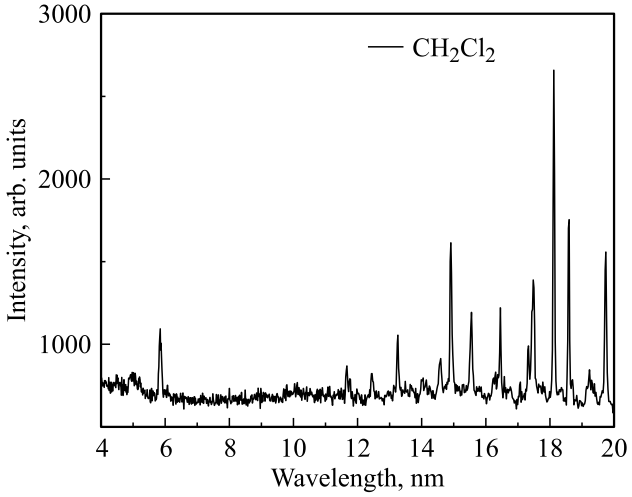

Figure 1. Emission spectra obtained with a liquid jet of dichloromethane CH_2Cl_2 used as the target.

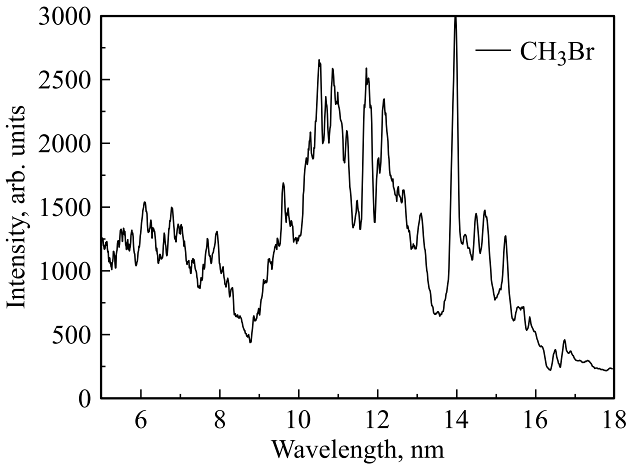

Figure 2. Emission spectra obtained with a liquid jet of methylene bromide CH_3Br used as the target.

processes, such as dispersion of a liquid jet, boiling of liquid in vacuum, and its freezing, proceed simultaneously in the course of outflow. Since the calculation of the structure of such a jet is a rather time-consuming process, it was not performed. However, it is certain that droplets (crystals) of liquid with a density on the order of $\sim 10^{24}$ mol/cm^3 are present in the laser spark generation region.

Results

Examination of hexane-based liquid targets

Hexane C_6H_{14} is a saturated hydrocarbon: transparent, thin, highly volatile, and low-toxic liquid with a low reactive capacity in the conditions of our experiment. The ease of evacuation, fine thermodynamic properties, and wide availability make hexane an excellent liquid target. As far as we know, hexane has never been used before as a target for an SXR and EUV LPS.

Table 1. Table of emission lines of CH_2Cl_2

Wavelength, nm	Intensity at maximum	Ion	Transition
5.0	800	Cl VIII	$2p^6-2p^53d$
5.85	1100	Cl VIII	$2p^6-3s$
11.66	860	Cl VII	$3s-7p$
11.76	790	Cl VIII	$2p^53s-2p^56p$
12.44	820	Cl VIII	$2p^53s-2p^55p$
13.31	1050	Cl VII	$3s-6p$
14.04	800	Cl VII	$3p-8d$
14.6	900	Cl VII	$3p-7d$
14.94	1600	Cl VII	$3s-5p$
15.58	1200	Cl VII	$3p-6d$
16.23	800	Cl VII	$3p-6s$
16.28	830	Cl VII	$3p-6s$
16.57	1200	Cl VIII	$2p^53s-2p^54p$
16.71	800	Cl VIII	$2p^53s-2p^54p$
17.4	1100	Cl VII	$3p-5d$
17.46	1400	Cl VII	$3p-5d$
18.25	2650	Cl VIII	$2p^53p-2p^54d$
18.63	1750	Cl VIII	$2p^53p-2p^54d$
18.78	780	Cl VIII	$2p^53p-2p^54d$
19.13	850	Cl VII	$3p-5s$
19.64	1550	Cl VII	$3s-4p$

The following parameters of liquid were set in the study of the hexane target: the pressure at the nozzle inlet was 4 bar, and the temperature was 300 K. No emission lines of carbon ions were detected in the 4–20 nm spectral range in these experiments. The reason for this is that the probability of emission of C V and C VI carbon ions produced under excitation of the hexane target in the 4–20 nm region is rather low; recombination radiation of carbon ions in the „water transparency window“ of 2.8–4 nm is significantly more probable. Hexane is a convenient carrier liquid that, as was verified experimentally, may be used as a solvent for the study of emission spectra of various chemical compounds.

Examination of liquid targets based on CH_2Cl_2

Dichloromethane CH_2Cl_2 is a transparent, thin, highly volatile, and low-toxic liquid with a low reactive capacity in

Table 2. Table of emission lines of CH_3Br

Wavelength, nm	Intensity at maximum	Ion	Transition
5.05	1300	Br X/Br VIII*	*
5.6	1500	Br X/Br VIII*	*
5.8	1400	Br X/Br VIII*	*
6.1	1700	Br X/Br VIII*	*
6.35	1600	Br X/Br VIII*	*
6.61	1500	Br X/Br VIII*	*
6.8	1700	Br VIII	$3d^{10}-3d^9 8f$
6.95	1700	Br VIII	$3d^{10}-3d^9 8f$
7.04	1600	Br VIII	$3d^{10}-3d^9 7f$
7.35	1200	Br VIII	$3d^{10}-3d^9 6f$
7.7	1350	Br VIII	$3d^{10}-3d^9 5f$
7.93	1400	Br VIII	$3d^{10}-3d^9 5f$
8.3	1100	*	*
9.08	950	Br VIII	$3d^{10}-3d^9 4f$
9.61	1900	Br X*	$3d^8-(3d^7 4p+3p^5 3d^9)$?
10.30	2200	Br X	$3d^8-(3d^7 4p+3p^5 3d^9)$
10.53	2900	Br IX/Br X*	$3d^9-3p^5 3d^{10}$ or $3d^8-(3d^7 4p+3p^5 3d^9)$
10.7	2400	Br X	$3d^8-(3d^7 4p+3p^5 3d^9)$
10.86	2900	Br IX	$3d^9-3d^8 4p$
11.21	2400	Br X	$3d^8-(3d^7 4p+3p^5 3d^9)$
11.48	1650	Br IX	$3d^9-3d^8 4p$
11.66	2600	Br IX	$3d^9-3d^8 4p$
11.75	3200	Br IX	$3d^9-3d^8 4p$
11.8	2700	Br IX	$3d^9-3d^8 4p$
12.01	2000	Br IX	$3d^9-3d^8 4p$
12.15	2500	Br IX	$3d^9-3d^8 4p$
12.65	1700	Br VIII	*
13.1	1600	Br VIII	*
13.89	2100	Br VIII	$3d^{10}-3d^9 4p$
13.99	3200	Br VIII	$3d^{10}-3d^9 4p$
14.25	1350	Br VIII	$3d^{10}-3d^9 4p$
14.5	1500	Br VII	*
14.72	1500	Br VII	$3d^{10}4s-3d^9 4s4p$
15.25	1300	Br VII	*

Note. * The emitting ions and/or transitions corresponding to these emission lines remained unidentified.

the conditions of our experiment. The ease of evacuation, fine thermodynamic properties, and wide availability make CH_2Cl_2 an excellent liquid target. As far as we know, dichloromethane has never been used before as a target for an SXR and EUV LPS.

Figure 1 shows the CH_2Cl_2 emission spectra measured using the injector with a single aperture 225 μm in diameter at the following parameters of liquid: the pressure at the nozzle inlet is 4 bar, and the temperature is 300 K. The 4–20 nm spectral range was studied. The radiation intensity is given in relative units. A series of intense lines formed by transitions of Cl VIII and Cl VII ions is seen in the 4–20 nm range. Lines of carbon ions are not observed in the studied spectral range. An enhancement of the noise component, which is attributable to the specifics of the spectral instrument used, is observed at short wavelengths. The absolute values of intensity of SXR and EUV radiation corresponding to this target are planned to be measured in subsequent studies.

The observed lines were interpreted in accordance with [5–9] by comparing the spectra measured with different targets. The obtained results are listed in Table 1 in the Appendix.

Examination of liquid targets based on CH_3Br

Methylene bromide CH_3Br is a transparent, thin, highly volatile, and low-toxic liquid, which is also a rather efficient solvent with a low reactive capacity in the conditions of our experiment. The ease of evacuation, satisfactory thermodynamic properties, and wide availability make CH_3Br a satisfactory liquid target. Methylene bromide has a drawback in that a jet outflowing into vacuum tends to freeze. As far as we know, methylene bromide has never been used before as a target for an SXR and EUV LPS.

Figure 2 shows the CH_3Br emission spectra measured using the injector with a single aperture 225 μm in diameter at the following parameters of liquid: the pressure at the nozzle inlet is 4 bar, and the temperature is 300 K.

The 5–18 nm spectral range was studied. The radiation intensity is given in relative units. A series of intense bands and lines formed by transitions of Br X, Br IX, Br VIII, and Br VII ions is seen in the 5–18 nm range. The spectrum agrees qualitatively with the one corresponding to krypton excitation. Lines of carbon ions are not observed in the studied spectral range. An enhancement of the noise component, which is attributable to the specifics of the used RSM-500 monochromator spectrometer, is observed at short wavelengths. The absolute values of intensity of SXR and EUV radiation corresponding to this target are planned to be measured in subsequent studies.

The observed lines were interpreted in accordance with [10,11] by comparing the spectra measured with different targets. The obtained results are listed in Table 2 in the Appendix. We failed to identify the transitions corresponding to a number of lines.

Conclusions

In the present study, the emission spectra of an LPS with liquid hexane C_6H_{14}, dichloromethane CH_2Cl_2, and methylene bromide CH_3Br targets were examined. These liquid-droplet targets were formed by spraying through an injector in a vacuum chamber. The obtained spectra were interpreted, and ions emitting in the examined spectral range were identified.

No emission lines of carbon were observed in the process of excitation of liquid-jet hexane targets. Therefore, hexane may be used in experiments as an inert carrier for dissolving halogens, chalcogens, and several other chemical elements in it and introducing them into the reaction volume.

Emission lines of Cl VIII and Cl VII ions were identified in the process of excitation of liquid-jet dichloromethane targets. It was found that dichloromethane may be used efficiently as a working liquid for an LPS.

Emission lines of Br X, Br IX, Br VIII, and Br VII ions were identified in the process of excitation of liquid-jet methylene bromide targets. It was found that methylene bromide is a partially serviceable working liquid for an LPS.

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Conflict of interest

The authors declare that they have no conflict of interest.

References

[1] A.N. Nechay, A.A. Perekalov, N.N. Salashchenko, N.I. Chkhalo. Opt. Spectrosc., **129** (2), 185 (2021).
[2] A.N. Nechai, A.A. Perekalov, N.N. Salashchenko, N.I. Chkhalo. Opt. Spectrosc., **129** (3), 363 (2021).
[3] A.N. Nechai, A.A. Perekalov, N.I. Chkhalo, N.N. Salashchenko. Opt. Spectrosc., **129** (7), 789 (2021).
[4] A.N. Nechay, A.A. Perekalov, N.I. Chkhalo, N.N. Salashchenko, I.G. Zabrodin, I.A. Kaskov, A.Ye. Pestov. J. Surf. Invest.: X-Ray, Synchrotron Neutron Tech., **13**, 862 (2019).
[5] NIST Atomic Spectra Database, Gaithersburg, 2009–2019. https://www.nist.gov/pml/atomic-spectra-database.
[6] C. Jupén, J. Fremberg, B. Fawcett. Phys. Scr., **30** (4), 260 (1984).
[7] C. Jupén. Phys. Scr., **36** (5), 776 (1987).
[8] H.G. Berry, J. Desesquelles, K. T. Cheng, R. M. Schectman. Phys. Rev. A, **18** (2), 546 (1978).
[9] V. Kaufman, B. Edlén. J. Phys. Chem. Ref. Data, **3** (4), 825 (1974).
[10] Y.N. Joshi, A. Nencioni, T.A.M. van Kleef. JOSA B, **2** (10), 1650 (1985).
[11] Y.N. Joshi, T.A.M. Van Kleef. Phys. Scr., **23** (3), 249 (1981).